# Thermoelectricity of $URu_2Si_2$: giant Nernst effect in the hidden-order state

Romain Bel[1], Hao Jin[1], Kamran Behnia[1], Jacques Flouquet[2] and Pascal Lejay[3]
*(1)Laboratoire de Physique Quantique(CNRS), ESPCI, 10 Rue de Vauquelin, 75231 Paris, France*
*(2)DRFMC/SPSMS, Commissariat à l'Energie Atomique, F-38042 Grenoble, France*
*(3)Centre de Recherche sur les Très Basses Températures(CNRS), F-38042 Grenoble, France*
(Dated: July 6, 2004)

We present a study of Nernst and Seebeck coefficients in the heavy-fermion compound $URu_2Si_2$, which hosts a phase transition of unsettled origin. A giant Nernst signal of unprecedented magnitude was found to emerge in the ordered state. Moreover, our analysis of the Seebeck and Hall data indicates that the ordering leads to a sudden increase in the entropy per itinerant electron and to a drastic decrease in the scattering rate.

PACS numbers: 74.70.Tx, 72.15.Jf, 71.27.+a



Among heavy-fermion superconductors, $URu_2Si_2$ is distinguished by the presence of a mysterious electronic order below $T_0$=17.5K[1, 2, 3]. A large amount of magnetic entropy ($S_{mag} \sim 0.2Rln2$) is lost in this phase transition[1]. Nevertheless, the intensity of the emerging magnetic Bragg peaks imply an anti-ferromagnetic order with a very weak magnetic moment($\sim 0.03\mu_B$/U)[4]. With the application of pressure, the the magnetic Bragg peaks intensify and a conventional AF ground state emeges[5, 6]. At ambient pressure, the small moment resolved cannot account by itself for the amount of entropy lost at the transition. However, ordering opens a sizeable gap in both spin and charge excitations. This unusual case of large macroscopic anomalies leading to a tiny magnetic moment has nourished an extensive investigation during the last two decades. Many proposals[7, 8, 9, 10, 11, 12] regarding the nature of what is now commonly called the hidden order of $URu_2Si_2$[13] have emerged. Some of these invoked exotic order parameters such as three-spin-correlator[7], quadrupolar ordering of localized moments[8] or an unconventional spin-density-wave[11]. In some others, weak antiferromagnetism is explained by considering the dual (i.e. localized and itinerant) character of the $5f$ electron in the context of a Fermi surface with nesting[9, 10, 14].

Several recent experiments indicate that the hidden order and Large-Moment-Anti-ferromagnetism(LMAF) are in competition[15, 16, 17]. Anomalies indicating a phase transition between two thermodynamically-distinct states have been reported in thermal expansion studies[16]. According to $^{29}$Si NMR measurements[17], the hidden-order state is spatially inhomogeneous with coexisting LMAF and paramagnetic regions. The apparent weakness of anti-ferromagnetism would be a consequence of the tiny fraction of the volume occupied by the magnetically-ordered electrons at ambient pressure. This result provides a strong case for an electronic phase separation between the two order parameters[18].

In this paper, we report on the thermoelectricity of $URu_2Si_2$ which provides new pieces to this puzzle. The hidden-order state was found to host a Nernst coefficient of an exceptionally large magnitude. This giant Nernst coefficient decreases smoothly with the application of a magnetic field and the extrapolation of data indicates that it would not survive the destruction of the hidden order at high magnetic fields[19]. On the other hand, the onset of the ordering is accompanied with an enhancement of the absolute value of the Seebeck coefficient. This is in sharp contrast with the concomitant decrease in the electronic specific heat[1, 3, 20]. We will argue that it can be understood by taking into account the change in the carrier density induced by the transition.

Single crystals of $URu_2Si_2$ were prepared in a three arc furnace under purified argon atmosphere and annealed under UHV for one week at 1050°C. Four longitudinal and two lateral contacts were made on each crystal. Nernst and Seebeck coefficients were measured using a one-heater-two-thermometers set-up which allowed us to measure thermal and electric conductivities as well as the Hall coefficient of the sample in the same conditions. The results reported here were reproduced on two different samples.

Fig.1 presents the temperature dependence of the Nernst effect for various magnetic fields in $URu_2Si_2$. The upper panel displays the ratio of the transverse electric field to the longitudinal thermal gradient (N=-$E_y/\nabla_x T$), while the lower panel presents the Nernst coefficient($\nu$=N/B). As seen in the figure, below $T_0 \sim$ 17.5K, the Nernst signal (which is negligibly small above this temperature) begins to grow steadily and attains its maximum value at T$\sim$3K. This Nernst signal presents an sublinear field-dependence as indicated by the field-induced decrease of $\nu$ seen in the lower panel of the figure. Plotting $\nu_{max}$ as a function of magnetic field, one can estimate the field scale associated with the destruction of the giant Nernst signal (see the lower inset of the figure). A simple linear extrapolation of $\nu_{max}(B)$ to higher fields puts this field at B$\sim$ 32T. Since this is comparable to the magnitude of the field required to destroy the hidden order($\sim$ 35T), it is safe to assume that the giant Nernst signal is another property of the ordered state.

The magnitude of the Nernst coefficient is remarkably large. In the zero-field limit, it attains $\nu_{max}$ $\sim$4.2 $\mu$V/KT, and easily exceeds what is reported in any other metal[21] including $NbSe_2$[22]($\nu_{max} \sim$ -0.12$\mu$ V/KT) or $CeCoIn_5$[23]($\nu_{max} \sim$ -0.95$\mu$ V/KT). At B=12T, the sig-

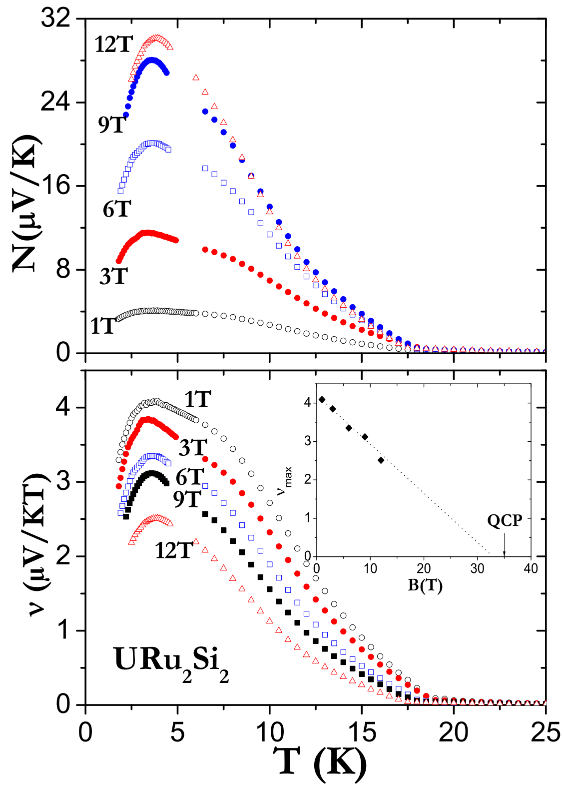


FIG. 1: Temperature dependence of the Nernst signal (upper panel) and coefficient (lower panel) for different magnetic fields in $URu_2Si_2$. Upper inset shows the temperature dependence of the transverse to longitudinal voltages produced by a longitudinal thermal gradient. Lower inset presents the field dependence of the peak value of $\nu$. The arrow presents the field corresponding to the destruction of the hidden order[19].

nal is even larger than what is observed in the vortex state of any superconductor. In the zero-field limit, however, the Nernst coefficient in a cuprate superconductor, $Ba_2Sr_{1.5}La_{0.5}CuO_6$ is reported to attain a comparable magnitude ($\nu_{max} \sim 4.5$ $\mu$V/KT)[24].

The phase transition which leads to the emergence of a Nernst signal of such a magnitude affects also the [longitudinal] thermopower[25]. As seen in Fig.2, the Seebeck coefficient, $S$, presents a sharp anomaly at $T_0$ and becomes strongly field dependent in the hidden order state. As seen in the inset of Fig.2, a new low-temperature maximum in $S(T)$ emerges in fields exceeding 6T. However, in order to resolve the magnitude of temperature-linear thermopower in the zero temperature limit, a plot of $S/T$ as a function of temperature[26] is more instructive. As seen in the main panel of the figure, the application of a magnetic field leads to a steady increase in the magnitude of $S/T$ down to the lowest temperatures investigated. In many metals, the absolute value of a dimensionless ratio, $q = \frac{S}{T\gamma} N_{Av} e$, linking the Seebeck coefficient to the elec-

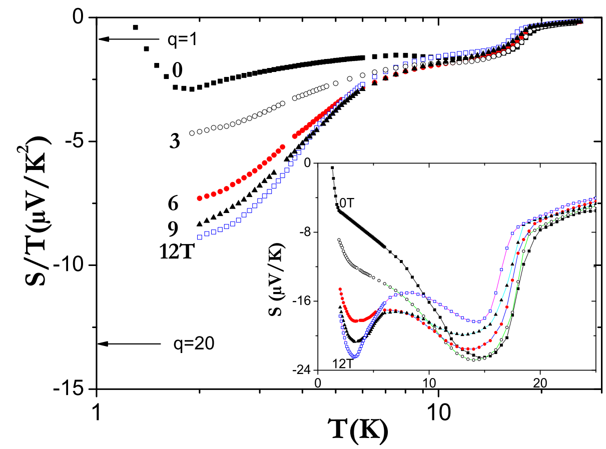


FIG. 2: Temperature-dependence of the Seebeck coefficient divided by temperature as a function of temperature for different magnetic field. The arrows point to the expected $S/T$ given the electronic specific heat of $URu_2Si_2$ for different carrier densities (see text). The inset shows the same data in a more traditional fashion.

tronic specific heat ($\gamma = C_{el}/T$) is close to unity[26] in the T=0 limit. [Here $N_{Av}$ is the Avogadro number and $e$ is the elementary charge.] This is not the case of $URu_2Si_2$. Taking the zero-field values of $S/T$ (-2.8$\mu$V/K$^2$) and $\gamma$ (60 mJ/molK$^2$)[20]) at the onset of superconductivity yields $q = -4.5$. The enhancement of the absolute value of $S/T$ with the application of the magnetic field (which leaves $\gamma$ unchanged in this range[20]) leads to an even larger $q$ at higher fields(q∼-14 at B=12T). The low carrier density in $URu_2Si_2$ provides a natural explanation for this large magnitude of $q$. The conversion factor, $N_{Av}e$, between thermopower and specific heat assumes that there is a single carrier per formula unit. Trivially, a proportionally larger $q$ is expected when the density of carriers is lower than this. The case for a small carrier density in the hidden-order state of $URu_2Si_2$ is supported by several other experimental observations. It can be directly deduced from the magnitude of the Hall coefficient in the zero-temperature limit which was found to be $R_H = 9.5 \times 10^{-3} cm^3/C$ in agreement with previous studies[27, 28]. While at finite temperatures, the magnitude and the temperature-dependence of the Hall coefficient of the Heavy-Fermion compounds depend largely on skew scattering[29], the extraordinary Hall effect vanishes in the zero-temperature limit. Assuming a one-band model, the magnitude of the Hall coefficient yields a carrier density of 0.05 holes per Uranium atom, as previously reported[27, 28]. Such a low carrier density implies a Fermi surface occupying a small portion of the Brillouin zone which is consistent with the results of band calculations based on the spin-polarized Dirac equation[30]. Direct evidence for small [and almost spherical] Fermi sur-

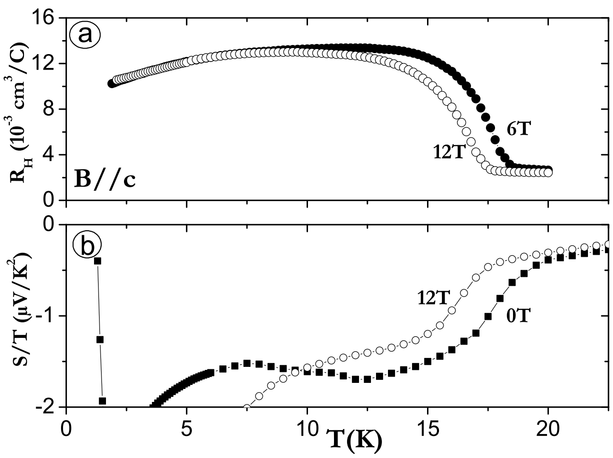


FIG. 3: a) Temperature-dependence of the Hall coefficient. b) Same for thermopower divided by temperature. Note the concomitant jumps in the vicinity of $T_0$.

faces comes from de Haas- van Alphen and Shubnikov-de Haas measurements[31, 32, 33]. In the most complete set of these studies, Keller *et al.*[33] identified four different orbits with the largest occupying less than 5 percent of the Brillouin zone.

Analysis of the behavior of thermopower and Hall coefficient in the vicinity of $T_0$ leads to a subtle reconsideration of what occurs to itinerant electrons at $T_0$. As seen in Fig.3 which compares the anomalies in $R_H$ and in $S/T$, the absolute value of both shows a step-like increase in a rather narrow temperature window($\sim$ 3K). The three-fold *increase* in the magnitude of $S/T$ contrasts with the concomitant *decrease* of $\gamma$ which passes from 180 mJ/K$^2$mol above $T_0$ to 60 mJ/K$^2$mol below. Therefore, the transition leads to a reduction of the entropy per volume yet an enhancement of the entropy associated to each carrier. This is confirmed by the sharp change in the Hall coefficient at $T_0$. The five-fold increase in $R_H$ corresponds to an even larger drop in carrier density since a sizeable fraction of the Hall signal at this temperature comes from skew scattering. Since the magnetic susceptibility is barely affected by the transition[27, 28], the contribution of skew scattering to Hall effect between 20K and 15 K remains unchanged. An analysis based on the Fert-Levy model[29] yields a carrier density of 0.4 holes per U above $T_0$ and ten times less below[28]. The three-fold decrease in $\gamma$ at $T_0$ appears, therefore, to mask two opposing tendencies: a ten-fold reduction in carrier density and a three-fold increase in entropy per carrier. In other words, while 90 percent of the carriers condense at the transition, those which survive carry a larger entropy.

Thus, instead of being a light-weight heavy fermion, $URu_2Si_2$ in the hidden-order state emerges from this analysis as a system with a very low density of carriers but a fairly large amount of entropy per carrier. A carrier density of 0.05 holes per f.u. implies $q \sim -20$. For $\gamma \sim 60$mJ/molK$^2$, this yields an expected $S/T = -12.5\mu$V/K$^2$. As seen in Fig.2, the low-temperature magnitude [of the absolute value] of $S/T$ at zero-magnetic field is considerably lower than this. With the application of magnetic field, $S/T$ is enhanced and approaches the expected magnitude without attaining it. Since $R_H$ (and consequently the carrier density) shows little variation with magnetic field in this range, the strong change in $S/T$ with magnetic field at finite temperature seems to reflect the field dependence of the scattering time.

Let us now consider the emergence of a giant Nernst signal in such a context. For this, it is useful to focus on a very simple relationship which relates the Nernst coefficient to the Hall angle in the Boltzmann picture and recently put forward by Oganesyan and Usshishkin[35]:

$$N = \frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{\partial \Theta_H}{\partial \epsilon}|_{\epsilon_F} \tag{1}$$

Since the Hall angle, $\Theta_H = \frac{\sigma_{xy}}{\sigma_{xx}}$, is a convenient measure of the scattering time, $\tau$ (namely, $\Theta_H = \omega_c \tau$, where $\omega_c$ is the cyclotron frequency), then, Eq. 1 states that the Nernst signal measures the energy-dependence of the scattering time at the Fermi energy. The latter quantity is small in common metals, leading to a negligible Nernst signal, a phenomenon also called Sondheimer cancellation[24]. The situation becomes more complicated in multi-band metals where the thermal gradient creates a counterflow of carriers with opposite signs creating a finite Nernst signal. Such an ambipolar Nernst effect was observed in $NbSe_2$ which happens to present a sign-changing Hall coefficient in its Charge Density Wave state[22]. The giant Nernst signal in $URu_2Si_2$, however, is more than one order of magnitude larger. Moreover, it emerges in presence of a large and *non-vanishing* Hall angle. In order to explore other possible sources of enhanced Nernst signal in a Boltzmann picture, let us make a crude approximation and replace the energy derivative $\frac{\partial \Theta_H}{\partial \epsilon}$ at the Fermi level by $\frac{\Theta_H}{\epsilon_F}$. In this case, Eq.1 yields:

$$N \approx 283\frac{\mu V}{K} \times \Theta_H \times \frac{k_B T}{\epsilon_F} \tag{2}$$

A number of elementary insights are provided by this simple expression. First of all, compared to simple metals, the heavy-fermion compounds are expected to display an enhanced Nernst signal simply because of the small magnitude of their Fermi energy. Experimentally, this is the case: in the two heavy-fermion metals studied until now, Nernst coefficient is found to be in the 0.2-5$\mu$V/KT range, several orders of magnitude larger than the isothermal Nernst coefficient of gold($\sim$ 0.1 nV/KT)[36]. In the second place, the expression indicates that any increase in $\Theta_H$ (or more fundamentally, in the quasi-particle lifetime $\tau$) would also lead to an enhancement of the Nernst coefficient. Interestingly,

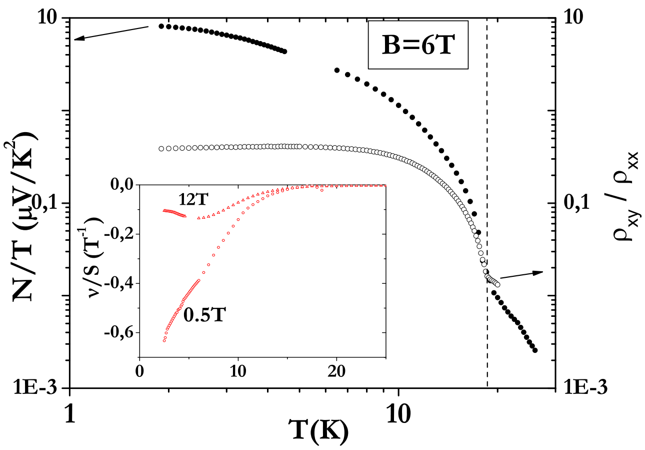


FIG. 4: Nernst coefficient divided by temperature and the Hall angle as a function of temperature. The dashed vertical line marks the transition temperature $T_0$.Inset: the temperature dependence of the ratio of the Nernst to Seebeck coefficients.

in both $URu_2Si_2$ and $CeCoIn_5$, the giant Nernst effect emerges with a concomitant enhancement in the amplitude of $\Theta_H$.

This brings us to another remarkable feature of electronic transport in the hidden-order state of $URu_2Si_2$. Fig.4 displays the temperature dependence of the Hall angle in a logarithmic scale. As seen in the figure, the phase transition at $T_0$ leads to an almost thirty-fold increase in the Hall angle. At B=12T, the system is close to the $\omega_c\tau \sim 1$ limit[34]. Following the thread of Eq.1, it is instructive to compare this with the temperature dependence of $N/T(T)$. As seen in Fig.4, both quantities are considerably enhanced below $T_0$. However, their behavior is not identical. When the sample is cooled from 17K to 1.5K, $\Theta_H$ increases by a factor of 30 and saturates rapidly. On the other hand, $N/T$ increases by a factor of 500 and continues to increase smoothly.

Therefore, a small Fermi Energy combined with a long scattering time provides a partial explanation for the magnitude of the Nernst coefficient in $URu_2Si_2$. Numerically, at B=6T and T=3K, $N \sim 20\mu V/K$ and $\Theta_H \sim 0.4$ and expression 2 implies $\epsilon_F \sim 17$K. However, a reduced Fermi energy would enhance the Nernst and Seebeck coefficients in exactly the same way. This is, however, not the case. As seen in the inset of figure, in the low-field regime, upon cooling the ratio $\nu/S$ displays a diverging behavior in a manner analogous to the one observed in $CeCoIn_5$[23]. In both cases, the large Nernst signal is accompanied with an [anomalously] small Seebeck signal. Moreover, as seen in Fig.4, $N/T$ continues to increase at low temperature while $\Theta_H$ has already attained a constant value. These features are yet to be understood. These features point to a missing ingredient for understanding this enhanced Nernst signal which may be a generic feature of heavy fermions physics.

We note that a link between unconventional density wave order and Nernst signal has been recently proposed[37]. On the other hand, given the experimental evidence in favor of electronic phase separation in the hidden order state of $URu_2Si_2$[17, 18], one is tempted to speculate in other directions. For example, magnetic droplets inside a paramagnetic fluid may couple a magnetic flux line to an entropy reservoir in an manner similar to superconducting vortices.

In summary, our results indicate that the ground state of $URu_2Si_2$ is one with a low density of itinerant electrons with large entropy and long lifetime. Most remarkably, we found that a Nernst signal of exceptionally large magnitude emerges in this context.


[1] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **55**, 2727 (1985)
[2] W. Schlabitz *et al.*, Z. Phys. B **62**, 171 (1986)
[3] M. B. Maple *et al.*, Phys. Rev. Lett. **56**, 185 (1986)
[4] C. Broholm *et al.*, Phys. Rev. Lett. **58**, 1467 (1987); Phys. Rev. B **43**, 12809 (1991)
[5] H. Amitsuka *et al.*, Phys. Rev. Lett. **83**, 5114 (1999)
[6] F. Bourdarot *et al.*, cond-mat/0312206 (2003)
[7] V. Barzykin and L. P. Gor'kov, Phys. Rev. Lett. **70**, 2479 (1993)
[8] P. Santini and G. Amoretti, Phys. Rev. Lett. **73**, 1027 (1994)
[9] A. E. Sikkema *et al.*, Phys. Rev. B **54**, 9322 (1996)
[10] Y. Okuno and K. Miyake, J. Phys. Soc. Jpn. **67**, 2469 (1998)
[11] H. Ikeda and Y. Ohashi, Phys. Rev. Lett. **81**, 3723 (1998)
[12] P. Chandra *et al.*, Nature, **417**, 831 (2002)
[13] N. Shah *et al.*, Phys. Rev. B **61**, 564 (2000)
[14] I. Fomin, to be published.
[15] M. Yokoyama*et al.*, cond-mat/0311199 (2003)
[16] G. Motoyama *et al.*, Phys. Rev. Lett. **90**, 166402 (2003)
[17] K. Matsuda *et al.*, Phys. Rev. Lett. **87**, 87203 (2001); J. Phys.:Condens. Matter **15**, 2363 (2003)
[18] P. Chandra *et al.*, J. Phys.:Condens. Matter **15**, S1965 (2003)
[19] K. H. Kim *et al.*, Phys. Rev. Lett. **91**, 256401 (2003)
[20] N. H. van Dijk*et al.*, Phys. Rev. B **56**, 14493 (1997)
[21] One possible exception is $(TMTSF)_2PF_6$ with a magnetic field oriented along a magic angle (See W. Wu *et al.*, Phys. Rev. Lett. 91, 056601 (2003)).
[22] R. Bel, K. Behnia and H. Berger, Phys. Rev. Lett.**91**, 066602 (2003)
[23] R.Bel *et al.*, Phys. Rev. Lett. 92, 217002 (2004)
[24] Y. Wang *et al.*, Phys. Rev. B **64**, 224519(2001)
[25] J. Sakurai *et al.*, Solid State Comm. **97**, 689 (1996)
[26] K. Behnia, D. Jaccard and J. Flouquet, J. Phys.:Condens. Matter **16**, 5187 (2004)
[27] J. Schoenes *et al.*, Phys. Rev. B **35**, 5375 (1987);
[28] A. L. Dawson *et al.*, J. Phys.:Condens. Matter **1**, 6817

(1989)
[29] A. Fert and P. M. Levy , Phys. Rev. B **36**, 1907 (1987)
[30] H. Yamagami and N . Hamada, Physica B **284-288**, 1295 (2000)
[31] C. Bergemann *et al.*, Physica B **230-232**, 348 (1997)
[32] H. Ohkuni *et al.*, J. Phys. Soc. Japan **66**, 945 (1997)
[33] N. Keller *et al.*, J. Magn. Magn. Mater.**177-181**, 298 (1997)
[34] In $URu_2Si_2$, quantum oscillations are easily observable at magnetic fields well below $H_{c2}$ in crystals with residual resistivities as large as $10\mu\Omega$ cm. This is a result of the unusual combination of the two factors discussed above, namely the low carrier density and the long quasiparticle lifetime.
[35] Oganesyan and Usshishkin, cond-mat/0312558
[36] R.Bel *et al.*, to be published.
[37] B.Dóra *et al.*, Phys. Rev. B **68**, 241102(R) (2003)